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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: François Pierre Michel CANSELL et al.
Title: METHOD FOR OBTAINING A COMPOSITE FERRO-ELECTRIC MATERIAL
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DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents
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Sir:

I, Cyril Gérard Jacques Aymonier, being duly warned, hereby declare and state:

1. I understand English when it is written.
2. I have a Doctor degree in Chemical Engineering from the University of Bordeaux (2000 – Supervisor: Pr F. Cansell). I carried out postdoctoral research with Pr R. Mülhaupt (in connection with ARKEMA company) and Pr S. Mecking (Freiburg, Germany, 2000-2002). I am currently employed as CNRS researcher (head of the team “Chemistry in supercritical fluids – 18 people) at the Institut of Condensed Matter Chemistry of Bordeaux (ICMCB) where I have been employed since 2002. I have been conducting research and development in the field of the chemistry in supercritical fluids for the synthesis of advanced nanostructured materials_for 6 years.

3. I am a co-inventor of the invention disclosed and claimed in U.S. Patent Application No. 10/575,472, which is a National Phase application of PCT/FR2004/002580.

4. I have carefully reviewed the disclosures of the following references:

- R. Chen et al., Materials Letters 54 (2002) 314-317 (“Chen”)
- U.S. Patent No. 4,552,786 to Berneburg et al. (“Berneburg”)

5. A person of ordinary skill in the field of coating ceramic powder reviewing Chen and Berneburg would have concluded that Chen discloses no clear evidence of the structure of the obtained material, that Chen and Berneburg solve different problems, that the method of Chen would not work under supercritical conditions.

Chen discloses no clear evidence of the structure of the obtained material

6. Chen discloses a method for making a ceramic material. According to Chen, the method comprises the steps of coating particles of barium titanate (BaTiO_3) with a layer of silica (SiO_2) by a sol-gel process and then sintering the coated particles by cold-pressing in order to form disk samples.

7. Chen explains that the coating is divided into two steps (see page 316, paragraph “3.2. Coating mechanism”). According to a first step, barium titanate particles are added into water, and a layer rich in TiO_2 is formed at the surface of the barium titanate particles (see paragraph “3.2.1. Slight hydrolysis of BaTiO_3 ”). Then according to a second step, the pH value of the solution is decreased by adding HCl, which causes gelation of SiO_2 onto the surface of the barium titanate particles (see paragraph “3.2.2. Na_2SiO_3 sol-gel process on BaTiO_3 ”).

8. The tests carried out by Chen on the powder and on the disk samples do not show that a core-shell structure is actually obtained. Figure 1 shows a particle as it can be seen using a high-resolution transmission electron microscope. Chen indicates : “From fig. 1, the homogeneous silica film can be seen clearly on the powder’s surfaces” (see page 315, paragraph “3.1. Characterization of the coated BaTiO_3 powder”). However, the so-called “silica film” which is visible on figure 1 could just as well be a layer rich in TiO_2 formed at the surface of the barium titanate particle, when the particles are added into water as

previously mentioned. Therefore, it cannot be concluded with certainty from Figure 1 that the method disclosed by Chen leads to barium titanate particles coated with silica.

9. Figure 2 discloses results of an energy dispersion spectrometry analyses. Figure 2 only confirms the existence of silica in the powder. However, it is not possible to deduce from Figure 2, that the powder is constituted of barium titanate particle coated with silica. It could be as well separate particles of barium titanate and silica.

Chen and Berneburg faced different problems

10. Berneburg solves a problem that Chen does not disclose. Berneburg seeks to fill pores inside a ceramic body. Berneburg's problem is "surface plugging and sealing-off of pores" at the surface of the ceramic body. Berneburg solves the problem of sealing-off of pores by applying a solution at supercritical conditions. The supercritical solution penetrates inside the ceramic to fill the volume of "interconnected internal pores." (See Berneburg, column 4, lines 14-16 and 30-32.)

11. Chen faces no problem comparable to the "surface plugging and sealing-off of pores" problem disclosed in Berneburg, because Chen merely seeks to coat the exterior surface of powder particles. These surfaces are fully exposed to the coating solution. Chen seeks to coat surfaces that are fully exposed and easily reached with coating solution.

Chen and Berneburg disclose nonequivalent processes

12. Depositing a precursor from a solution within the interior of pores (as in Berneburg) is not an equivalent physical process to precursor deposition by a chemical way onto the exterior of particles (as in Chen).

13. Deposition into ceramic pores requires penetration of pore channels by the solution from surface openings into the interior of the ceramic. The surfaces to be coated are unexposed, within the ceramic. This introduces problems of pore clogging, which prevents full penetration into the ceramic and reduces exposure to fresh, unreacted solution for interior pores.

14. Precursor deposition onto ceramic particles is much simpler than coating interior pores of a ceramic body, because pore penetration is unnecessary for deposition onto ceramic particles. Particle surfaces to be coated are fully exposed.

15. In Berneburg, a low density ceramic body is infiltrated with a supercritical fluid containing a ceramic precursor. Berneburg states : “It has been discovered that the highly permeative nature of supercritical fluid allows for much easier infiltration (penetration) of smaller openings in ceramic material and can thus improve the densification of ceramic bodies.” (Berneburg, column 2, lines 12-16). In other words, Berneburg discloses that supercritical conditions enhance infiltration in submicron sized pores.

16. Berneburg does not disclose or suggest that supercritical conditions enhance “precipitation.”

The method as disclosed by Chen would not work under supercritical conditions

17. The Office Action dated July 7, 2008, states “*However, as discussed in the previous office action and in greater detail above, it would have been obvious to one of ordinary skilled in the art to modify Chen in view of Berneburg in order to carry out the Chen process with a supercritical fluid. Chen teaches an aqueous solution of the dielectric precursor, and a supercritical water solution is at pressure well above 10 bar. Therefore, the obvious modification of Chen et al in view of Berneburg et al would lead to a process meeting all of the limitations of the newly amended claim 1.*” (see pages 4-5, paragraph 5 of the Office Action).

18. However, the quoted analysis in the previous paragraph is technically incorrect for at least two reasons.

19. The first reason is that Chen’s method would not work as intended under supercritical conditions.

20. NaSiO_3 is nearly insoluble in supercritical water (water above 374°C and 221 bars). So it is impossible to coat the barium titanate particles with silica according to the method of Chen.

21. The second reason is that Berneburg supercritical fluid is incompatible with Chen's method.

22. The method of Berneburg is not suitable for coating particles as disclosed in Chen. In Berneburg, there is no chemical reaction between the precursor and the ceramic body. The precursor is simply "trapped" into the pores of the ceramic body while the fluid (propane) escapes from the ceramic. To the contrary, Chen requires specific chemical reactions between the barium titanate and the silica precursor in order to form a coating layer over the particles.

23. Berneburg's process is not designed for depositing the silica coating of Chen. Silica would not dissolve in the Berneburg supercritical fluid.

24. In Chen's process one could not replace Chen's fluid, water, with Berneburg's fluid, propane. Berneburg's fluid propane cannot superficially dissolve barium titanate in order to form TiO_2 (see Chen page 316, paragraph 3.2.1.). Moreover, propane is not able to dissolve HCl, which is needed as a catalyst for controlling pH and releasing a chemical reaction between superficial TiO_2 and silica (see Chen page 315, paragraph 2.1.). Therefore, supposing that the method of Berneburg was used for infiltrating particles of barium titanate (instead of a porous ceramic), this would lead to obtaining a blend of barium titanate particles and initial silica precursor, because there would be no possible chemical reaction between the compounds.

25. If a person of ordinary skill in the art tried to modify the process disclosed by Chen by applying the teaching of Berneburg, he would rather make a porous ceramic of barium titanate and then infiltrate the porous ceramic with a supercritical fluid containing silica precursor, in accordance with Berneburg. The resulting material having a completely different structure from that of our invention as claimed in patent application 10/575,472. In particular, the resulting material would not be ferroelectric particles that are isolated from one another by a dielectric layer.

26. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like

so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent resulting therefrom.



13/10/08

Date

Cyril Gérard Jacques Aymonier